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A BRIEF REVIEW OF PLANNED R&D IN THE LIMB DEVELOPMENT PROGRAM

by

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16. ABSTRACT The paper is a brief review of work planned and work underway in support of the Limestone Injection Multistage Burner (LIMB) development effort. A detailed description of the equipment used in the Innovative Furnace in-house effort is given. Work plans for contracts and a cooperative agreement with Acurex Corporation, Energy and Environmental Research Corporation, and Southern Research Institute covering internally staged burner work, fundamental combustion research, staged fuel NOx reduction, tangential-fired furnace pilot project, and LIMB particulate emissions are reviewed. The in-house work on limestone properties and reactivity and sorbent properties/fundamental chemistry is mentioned. The planned "super sorbents" and graphite drop-tube furnace work are covered. No results are discussed. Mention is made of lime-based sorbents with surface area of over 60 sq m/g as measured by a B.E.T. unit using N2 as the absorbed gas. Tests on furnaces ranging in size from 2.9 W to 29 MW are planned. Particle size distributions of total furnace emissions, before control devices, in the 0.1 micrometer range are to be measured. Reduction of both SOx and NOx is one goal of the effort.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group	
Pollution	Nitrogen Oxides	Pollution Control	13B	
Coal	Particles	Stationary Sources	21D	14G
Natural Gas	Dust	Limestone Injection		11G
Burners	Graphite	Multistage Burners	13A	08G
Calcium Carbonates		Particulates	07B	
Combustion	Calcium Oxides		21B	

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INTRODUCTION

This brief review will cover some of the efforts or projects planned for the Limestone Injection Multistage Burner (LIMB) Development Program. As an example of an in-house project, a detailed survey will be made of the principal equipment used in one project, the innovative furnace in the Wing "C" laboratory of the U.S. Environmental Protection Agency's Industrial Environmental Research Laboratory, Research Triangle Park, NC.

The scope of work for those projects not yet awarded will be used as a basis to describe them, and the work plan will be used as a basis to describe those projects underway.

DISCUSSION OF PROJECTS

EPA Innovative Furnace

The EPA innovative furnace is a modified copy of a furnace built by Energy and Environmental Research Corporation (EERC). The unit is a 6-in. (155-mm) internal diameter down-fired unit. Three burner configurations may be used: an axial-fired, a radial-fired, and a premix configuration with provision for tangential or swirl air. Each of the three configurations may employ the distributed-mixing burner concept. In the premixed burner this is accomplished by mixing less than the total air in the burner and adding the remainder of the air after the burner.

The firing rate of the furnace is adjustable with a nominal rating of 100,000 Btu/hr or about 3.2 kg coal/hr. Stable operation at a rate of about 1.4 to 1.6 kg coal/hr (42,000 to 50,000 Btu/hr) is a normal mode for testing. The unit may be fired on either propane or coal. Normal practice is to fire the unit on propane, unattended, over weekends, Friday afternoon to Monday morning, and overnight on weekdays, 1730 to 0800 hours. This allows thermal stabilization of the unit and avoids shock to the refractory by maintaining the unit at the testing temperature.

The unit is of conventional construction with rolled steel rings 3 ft (915 mm) in diameter and 1 ft (300 mm), 1.5 ft (450 mm), or 2 ft (600 mm) high. The steel rings are insulated by four courses of refractory beginning on the outside with a good insulating but low strength castable block mix refractory, progressing inward with castable insulation, following with Casolite 30,* and ending on the inside with a high strength, erosion resistant "hard face" refractory such as Green Cast 97. After casting the refractory into the steel rings, they are "stacked" using a gasket made of Kaowool or similar material on the outer periphery. A small amount of uncured refractory is applied around the matching faces of the inner central passage to prevent gas migration to the outer steel shell. Both the gasket and the uncured inner seal material are formed and spread by the weight of the ring and the clamping force of the bolted ring flanges, thus ensuring a good seal at each joint. The joint surfaces of the combustion passage are "faired in" and troweled to give a reasonably smooth and uniform combustion gas passage.

Inspection or view ports are provided, one a 2-in. (52-mm) circular port at about 7-1/2 in. (190 mm) from the burner tip, and a rectangular port 6 in. x 4 in. (155 mm x 100 mm), centered 23 in. (585 mm) below the burner tip. Nine thermocouples, embedded in the refractory approximately 1/2-in. (12 mm) from the combustion gas central passage, are spaced over the 11-ft (30.48-cm) long furnace section. There are eight ports, usable for either sampling or injection, approximately opposite the thermocouple locations. Three larger rectangular ports are available for the insertion of chokes and cooling coils to "stage" the combustion or divide the furnace into two or three zones with varied stoichiometry; the first is usually a "fuel-rich" or "reducing" zone, and the last, an air-rich or burnout zone.

Solid fuel, such as coal, is metered from a K-Tron™ loss-in-weight feeder. A similar feeder, but suited to delivery of a smaller amount, meters the sorbent, such as limestone or lime. The material is aspirated and transported by air and injected into the furnace through a water-cooled probe.

Operating conditions and emissions are monitored using the following instruments:

Digistrip III™ Data Logger
Continuous Monitors:

CO	Beckman Model 864 Infrared
CO_2	Beckman Model 864 Infrared
O_2	Beckman Model 755
SO_2	Anacon Model 207 U.V.
NO, NO_2, NO_x	Thermoelectron Model 10AR
HC	Beckman Model 402

Thermocouples, both B&K types, embedded or sealed in the refractory.

(*) Mention of trade names or commercial products does not constitute endorsement or recommendation for use by the U.S. EPA.

In addition, gas temperatures are measured using suction pyrometry with stabilized run conditions equal to those in the tests. No significant temperature difference was found between gas firing and coal firing at equal heat input rates, based on fuel heating value, and excess air conditions. The actual gas temperatures for different positions at the test conditions are thus established, and no conversion from the temperatures read by embedded thermocouples to the gas temperature is required. The relationship between thermocouple temperature at a given point under known conditions, and the gas temperature is known from the previous suction pyrometry work. Periodically, suction pyrometry tests are performed during emission testing to confirm the gas temperatures found in earlier suction pyrometry testing.

The SO_2 readings and NO_x readings from the continuous monitors are compared with those from EPA Methods 6 and 7 (wet chemistry). It would appear from preliminary results that the SO_2 readings are within 10 percent of the values of EPA Method 6 but that the NO_x readings are more than 10 percent from the values of EPA Method 7. This is being investigated further. Some difficulty (leaking glass-to-glass seal on the impinger vials) was encountered in collecting Method 7 samples.

Samuel L. Rakes is project officer for EPA Contract 68-02-3943 (with Northrop, Inc.) which provides a staffing level of one engineer and two technicians for this effort.

In-House Support Contract

This effort is now in the bid evaluation/negotiation stage. This contract will consolidate the support provided by contractors, in the medium of personnel qualified to operate the various pieces of equipment and laboratory apparatus, under one contract versus the three or more now covering the effort. The required level of training, qualifications, and number of persons required at each level are specified, and the offerors submit bids to provide this level of effort in support of the laboratory effort.

Sorbent Properties--Fundamental Chemistry

This work has provided activation energies for the lime/limestone and sulfur reactions, has provided data for kinetic plots versus surface area, and has explored the effect of calcination gas-phase compositions and temperatures for calcium-based materials. The work will be continued to extend the range of surface area reaction rates included in the data base, to explore the reaction kinetics, and to verify and expand the data base collected to date. It will be staffed under the in-house support contract at the level of three to four persons. The work will be under the technical direction of Robert H. Borgwardt.

High Temperature Reactions, Graphite Drop-Tube Furnace

This effort is intended to extend the temperature range in which fundamental lime/limestone and sulfur species reactions are studied to about

1800°C. The plans also call for sorbent and coal ash interactions to be studied at temperatures more nearly approaching flame temperatures seen in distributed-mixing burners. The equipment can handle a wide range of gas mixtures or atmospheres including highly flammable mixtures such as those containing hydrogen.

The troublesome and not-well-understood sorbent/coal ash interactions are scheduled for study in this furnace using a matrix derived from previous data in the sorbent properties fundamental chemistry effort, projections of these data, and data from the limestone reactivity effort. This effort will be staffed by a degreed professional under the in-house support contract. Technical direction will be by David A. Kirchgessner and Samuel L. Rakes.

Limestone Properties/Reactivity

This ongoing effort employs a variety of tests to determine the properties and reactivities of limestone and lime. An optical microscope, a mercury porosimeter, and a Brunauer-Emmett-Teller (B.E.T.) unit using N_2 to determine surface area, are used to develop information useful in determining those properties of limestone/lime that indicate or affect reactivity as related to SO_2 or sulfur capture reactions. In addition, there is a Dionex ion chromatograph (IC) to determine anion species and an atomic absorption (AA) unit to determine cation species.

This project has taken the techniques and conditions of calcination shown to give very high, up to 90 m^2/g, surface areas in lime when used for dispersed, milligram quantities and applied the principles to 250-plus g quantities. The techniques used have included fluidizing with nitrogen during calcination and during post-calcination cooling. The fluidizing technique has yielded surface areas higher than non-fluidized techniques; i.e., from about 20 m^2/g up to 50 m^2/g for those samples fluidized. The investigation will continue to search for a means to replicate the up to 90 m^2/g surface area (B.E.T.) seen in milligram quantities of limestone calcined in flowing N_2 while dispersed on quartz wool. This effort also includes geological evaluation and interpretation of the grain size and structure and crystal lattice type. David A. Kirchgessner is technical director/project officer for this project, staffed with two persons under the in-house support contract.

Superactive Sorbents

This is a new procurement, now in the bid evaluation/negotiation phase. The thrust of this procurement is to evaluate available data and devise a technique that will produce high surface area (40 m^2/g or better by B.E.T.) sorbents on a commercial scale; i.e., about 225 kg/hr. The project is planned in phases with the final phase being production of about 220 kg of the high surface area sorbent for use in a unit with a nominal firing rate of about 35 to 50 kg coal/hr. In this procurement, surface area has been equated to activity, since surface area is the easier parameter to measure and has in the past shown a direct correlation with reactivity.

Two conceptual designs are to be presented. One of the designs will be selected and carried through a detailed design study with a flow sheet, engi-

neering cost estimates, and cost-per-pound of sorbent delivered to the boiler prepared and presented. The 225 kg of sorbent for testing is to be prepared using, as closely as possible, the process described in the detailed design study.

Two approaches to production of the superactive sorbent are considered: in one, the sorbent is prepared and stored before use; and in the other, the sorbent is prepared immediately before injection into the boiler. In-situ preparation in the flame is not within the scope of the procurement. Samuel L. Rakes is the project officer for this procurement.

Tangential-Fired Furnace Pilot Project

This work is being done at Acurex Corporation, on a 439-kW (1.5×10^6 Btu/hr) tangentially fired pilot furnace. There are some 11 major components of the contract, called the "T-Firing Contract" for short, since the work is to investigate the parameters of the tangential- or T-fired boiler units.

The first major component was to confirm that the bulk-gas time/temperature profile through the pilot unit proposed for use in the work was comparable to that of the commercial tangential furnaces. Once this was accomplished, cold flow tests were performed to determine velocity profiles, actual gas residence time distribution (as opposed to bulk residence time), and particle residence time distribution. These results were extrapolated to full combustion temperature. Bulk temperature and gas composition information for coal firing with no sorbent for two furnace operating conditions was verified. Flow pattern information was also checked and verified as being acceptable. The bulk gas time/temperature profile during natural gas combustion at the firing conditions anticipated for coal was determined. Hydrogen sulfide (H_2S) was added to the gas, and the SO_2 concentration profile through the pilot unit was determined. This profile was compared to the SO_2 concentration profile observed during coal firing.

The sorbent activation profile while burning natural gas and the effectiveness of the distribution of the sorbent across the furnace were determined. This included particulate sampling traverses at various locations in the furnace. These samples were stored under nitrogen and shipped to North Carolina for analysis, including measurements of surface area, pore size distribution, and particle size distribution. This information on the sorbent activation profile, with the knowledge of the time/temperature profile and the SO_2 concentration profile mentioned previously, will be applied to a mathematical model for sulfur capture using sorbent injection during the firing of gas to which H_2S has been added. Tests will then be conducted using the gas with H_2S added to compare predicted results with the actual values observed.

The comparisons between the model predictions and the observed values will be analyzed. This analysis will indicate if changes in the model are needed.

The effects of coal ash deactivation or interference with sulfur capture are to be studied using a gas-fired regime with sorbent injection and with actual coal ash, or synthetic material to simulate coal ash, being injected at the normal coal injection points and in the usual amount seen when firing coal.

Solids samples would be analyzed as mentioned earlier. In this case, additional analyses (e.g., Auger microprobe) would be conducted in an effort to define the nature of any interaction between the sorbent and the coal ash material.

A final item in the project will be to inject pure $CaSO_4$ through the fuel injectors while burning natural gas with no sorbent. The solids leaving the furnace would then be analyzed to determine the degree of decomposition of the calcium sulfate in the furnace. The stability of the sulfate in the furnace should provide an indication of the degree to which sulfur capture by sorbent might be affected by sulfate stability.

The experimental operation is nearing completion, but data analysis and evaluation are pending. D. Bruce Henschel is project officer for this effort.

Internally Staged Burner Project

This work, covered by Contract 68-02-3923 with the Energy and Environmental Research Corporation, has been underway for approximately 2 years. The project officer is D. Bruce Henschel. The objective of this program is to develop generic design criteria for low-NO_x and low-SO_x burners which can be effectively retrofitted to existing wall-fired coal boilers. Prior efforts to retrofit externally staged low-NO_x burners (with outboard tertiary air ports) have shown that the large throat and the external tertiary ports of such burners can be difficult to accommodate in an existing boiler without significant modifications; hence, the desire to develop burners which can achieve equivalent staging effects internally, have characteristics that allow easy burner retrofit, and at the same time permit sorbent injection in a manner which enables effective SO_2 control.

The first phase of this study includes broad parametric testing at a 2.9 MW (10×10^6 Btu/hr) scale to screen the effects of a variety of design and operating variables on several candidate burner designs. These candidate burners would be selected following an assessment of retrofit requirements and vendor state-of-the-art. The goal would be to identify the burner designs and variables which provide the most effective NO_x control, SO_x control, and combined NO_x/SO_x control under conditions amenable to retrofit.

The next phase is a detailed characterization and more complete parametric studies including the burner(s) and the burner conditions selected using the information from the first-phase parametric screening. Measurements of gas species, solid species, and temperature are to be be made as a function of residence time. The slagging and fouling properties and in-situ resistivity are identified as special points to be checked regarding any change in fly ash properties during the tests. Testing will be on scales from 2.9 W to 29 MW (10 Btu/hr to 100×10^6 Btu/hr).

Since sorbent injection for SO_x control during combustion is a key element in developing design criteria for a retrofittable low NO_x/SO_x burner, a significant part of this effort will involve definition of sorbent injection criteria which will provide effective sorbent activation, good dispersion, and maximum residence time at sulfation reaction temperatures.

The product of this effort is to be a generic approach to burner design. This includes prediction of the optimum system at full scale. An alternate system, although not optimum but better understood and ready for large-scale testing, is also to be proposed.

Fundamental Combustion Research

This effort has four major objectives for 1984. The first of these is to measure surface area, pore size distribution, and sorbent reactivity as a function of time exposed to flame or near-flame temperatures and to apply the information gained to explore the ability to generate high-surface-area, highly reactive, sorbents using in-situ calcination within the furnace system. The partial pressures of the chemical species expected to be present, including CO_2, H_2O, and SO_2, will be varied from zero to the value expected based on measurements made in several furnaces. Particle heating rate will be investigated as a parameter affecting ultimate surface area.

The second major objective is to explore the application of high-surface-area sorbents, generated external to the furnace. The surface area decay rate for such precalcined sorbents, after injection into a furnace environment, will be measured. The same variables that were investigated for the in-situ calcination material are to be checked here. The change in sorbent properties while the externally precalcined sorbent is in storage will be investigated.

A search will be conducted for a technique and injection location that will avoid the observed coal ash deactivation of the sorbent. The mechanism and nature of the ash/sorbent interaction will be investigated in an attempt to determine the cause(s) of the observed deleterious effects of coal ash on sorbent activity. The investigations will include attempts to quantify the "stickiness" of ash and sorbent under a range of combustion conditions.

The coal will be divided into test fractions to obtain data on the probable usefulness of coal cleaning to remove those materials in the coal which are the greatest contributors to ash deactivation of the sorbent. Microphotography and microprobe analysis of collected particles will be used to explore the mechanisms of coal ash deactivation of the sorbent.

All of the fundamental work will be conducted on small laboratory combustion reactors. D. Bruce Henschel is project officer for this effort.

Staged Fuel NO_x Reduction

This technology, which has also been called "in-furnace NO_x reduction" and "staged fuel NO_x reduction," has been developed by Japanese boiler manufacturers for gas- and oil-fired systems, and limited work has been done on coal. In contrast to the conventional combustion process where all of the fuel is introduced through the burners, in the "reburning" process for NO_x control, only part (about 80 to 90 percent) of the fuel is fed to the primary combustion zone. The remainder (10 to 20) percent is injected following the primary combustion zone, creating a reducing "reburning zone." Conditions in the reburning zone determine the percentage reduction of NO_x. The reburn-

ing zone is in turn followed by an "oxidation zone," in which further air is added to complete the combustion of the "reburning" fuel. The Japanese work has concentrated on NO_x reduction only. EPA is conducting further investigations to determine if the reburning process offers any advantage for SO_x control by sorbent injection.

Under Contract 68-02-3925 with EPA, Energy and Environmental Research Corporation is conducting a study to better understand the reburning process, with emphasis on pulverized coal, and to optimize it for both NO_x and SO_x control. An initial phase of testing has been completed on a 29 kW (100,000 Btu/hr) down-fired furnace, providing information on reburning performance over a wide variety of operating variables. Using this information, the process will now be tested on a 34 MW-102 MW (10-30 x 10^6 Btu/hr) scale.

Capital and operating costs for the staged-fuel-NO_x reduction technique when used on units with rated electrical power output of 50, 250, 500, and 1,000 MW are to be estimated based on the larger scale tests. From this information, recommendations for large pilot-scale tests are to be made. All suitable boiler designs are to be included and potential problems identified. Some boiler designs do not have adequate space above the burners, some have a membrane-type wall which is not amenable to the perforations needed to introduce the secondary fuel, and others have similar design-specific features, such as fluid flow patterns, which cause them to be unsuitable for the "staged-fuel-NO_x reduction" technique. D. Bruce Henschel is project officer for this effort.

Pilot-Scale Evaluation of LIMB Particulate Emissions

This evaluation involves a cooperative agreement with Southern Research Institute. James D. Kilgroe is project officer for the agreement. This agreement is beginning the third year of effort. The final report is expected in mid to late 1985.

Briefly, the research program objectives can be stated in four parts:

1. Evaluate the effect of LIMB technology on particulate characteristics and existing particulate control system (electrostatic precipitator only) performance.

2. Evaluate the effect of sorbent injection on fouling of boiler tube surfaces.

3. Expand the existing data base concerning the capability of LIMB for meeting NO_x and SO_2 emission reduction goals under various circumstances.

4. Identify optimum integrated air pollution control system configurations incorporating LIMB technology.

Phase I, familiarization, shakedown, and verification of equipment operation, is completed. Phase II, the development of a baseline-data set using the Southern Research Institute equipment and instrumentation with coal and sorbent tested elsewhere, is nearing completion.

The Phase III effort plans detailed studies of five areas:

1. Electrostatic precipitator (ESP) rapping.

2. Effects of sorbent addition on the particle size distribution of the total particulate emissions, before any control device.

3. Fouling.

4. Residence time distribution (RTD).

5. Particulate characterization.

The first area, ESP rapping, will investigate the effect of sorbent addition on the tenacity of the particulate deposits collected on ESP electrodes. The force required to dislodge the collected particulate using standard rapping procedures will be determined using calibrated accelerometers mounted on the electrodes.

The particle size study will document the effect of sorbent addition on particulate size distribution (PSD) using a selected coal and sorbent combination. The PSD will be measured using an electrical aerosol size analyzer (EASA), an optical counter (OPC), and a cascade impactor. Previous studies found no sorbent effect on PSD below about 0.1 μm but did show an increase, by a factor of 3 to 4, in the particulate mass concentration in the range of 0.1 μm to 0.6 μm. The effects of different coals, different sorbent injection modes, and different sorbent types will be studied in additional tests of a less detailed nature in Phase III.

The fouling studies will assess the effect of sorbent addition on the fouling tendency of the particulate matter on boiler tubes. Both rate of deposition and strength of the deposits will be evaluated with and without sorbent addition. The probes will be air cooled to simulate the tube metal temperatures found in actual boiler tubes.

The strength of the deposits will be assessed using the Barnhart and Williams sintering test [see Transactions of the American Society of Mechanical Engineers (ASME) 78, 1229-1236, August 1956]. Current plans call for two coals to be tested with the sorbent injection modes to be selected through consultation of EPA and the Southern Research Institute.

The residence time distribution (RTD) studies are intended to provide a more accurate representation of the flow patterns and time/temperature relationships in the pilot combustor. Hopefully, some of the differences in results obtained between nominally identical equipment at different test facilities can be resolved. The gas flow pattern will be mapped with a stimulus/response technique using pulse injections of the radioactive tracer gas Krypton-85. The results should indicate how variations in flow patterns and time/temperature profiles can affect SO_2 capture.

Studies will be conducted using a scanning electron microscope (SEM), a transmission electron microscope (TEM), and X-ray diffraction (XRD) instruments to physically characterize the sorbent and the ash/sorbent mixture. SEM has been useful in examining the morphology of calcined and sulfated sorbent particles. Particle sections are prepared on a microtome and examined under TEM to investigate the internal structure of the particles. XRD scans for calcium, sulfur, silicon, and aluminium are used to identify the composition of sorbent and fly ash particles. Studies so far indicate that most of the sorbent particles become sulfated, but a small fraction escape with little or no sulfation. This last observation has been made consistently in sorbent samples generated with SO_2-doped natural gas. It is hoped that the XRD technique can be developed to identify specific compounds [e.g., CaO, $CaSO_4$, $CaCO_3$, $Ca(OH)_2$].

SUMMARY

This paper provided an in-depth review of one in-house project conducted in support of LIMB technology and a brief overview of some of the other projects underway or projected for the support of the LIMB program.